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Commercial Development of the ADVACATE Process for Flue Gas Desulfurization

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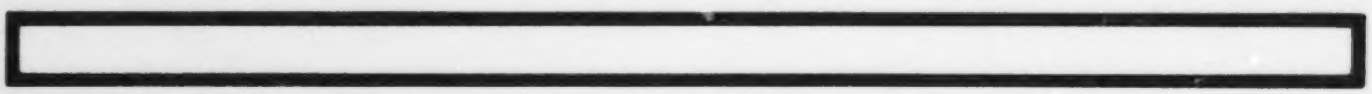

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16. ABSTRACT The paper discusses the commercial development of the advanced silicate (ADVACATE) process for flue gas desulfurization (FGD). This commercialization follows several years of bench-scale sorbent development, pilot plant process optimization, and field pilot evaluation. Results are presented showing the importance of silica dissolution in preparation of calcium silicates. Pilot and field optimization and evaluation shows excellent potential for over 90% SO_2 control at half the cost of conventional wet FGD processes. Projected costs and commercialization plans are presented.

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COMMERCIAL DEVELOPMENT OF THE ADVACATE PROCESS FOR FLUE GAS DESULFURIZATION

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ABSTRACT

Commercialization of the ADVACATE process for flue gas SO_2 removal follows several years of bench-scale sorbent development, pilot plant process optimization, and field pilot evaluation. Results are presented showing the importance of silica dissolution in preparation of calcium silicates. Pilot and field optimization and evaluation shows excellent potential for over 90 percent SO_2 control at half the cost of conventional wet flue gas desulfurization (FGD) processes. Projected costs and commercialization plans are presented.

SORBENT DEVELOPMENT

Under U.S. Environmental Protection Agency sponsorship, Jozewicz and Rochelle [1] studied the reaction of flyash with calcium hydroxide [$Ca(OH)_2$] for the production of reactive sorbents for use in FGD. They used the bench-scale packed bed reactor shown in Figure 1 which is capable of simulating conditions in the baghouse. They found that solids produced by slurrying flyash and $Ca(OH)_2$ were significantly more reactive than $Ca(OH)_2$ alone. The conversion of $Ca(OH)_2$ [defined as the number of mols of SO_2 reacted with 1 mol of $Ca(OH)_2$] was proportional to the weight ratio of flyash to $Ca(OH)_2$. The results of experiments at 54 percent relative humidity are presented in Figure 2. Increasing the flyash-to-$Ca(OH)_2$ weight ratio from 0.5 to 20 increased $Ca(OH)_2$ conversion from 0.17 to 0.78. For a fixed flyash-to-$Ca(OH)_2$ weight ratio, the slurrying temperature was the decisive parameter affecting the process. There was a critical slurrying time for every temperature tested, for which conversion reached a maximum value. The time needed to reach this maximum shortened with increasing temperature. For example, at a flyash-to-$Ca(OH)_2$ weight ratio of 16, $Ca(OH)_2$ conversion converged on 0.4 after 16 hr of slurrying at 25 °C and on 0.8 after 5 hr at 92 °C. This was a dramatic improvement of $Ca(OH)_2$ conversion, compared with 0.12 for $Ca(OH)_2$ alone at 54 percent relative humidity. The above work concluded that silica was the most reactive compound of flyash and that the reactivity increased with slurrying time and temperature. It was postulated that the rate limiting step of the reaction of flyash with $Ca(OH)_2$ was the dissolution of silica from flyash.

Several approaches were taken to increase the dissolution rate of silica. In one series of experiments, different chemical species were added in varying concentrations to a solid mixture at a 3:1 flyash-to-$Ca(OH)_2$ ratio. The most significant enhancement was seen when 0.25 N NaOH was added to the sample slurried for 8 hr at 90 °C where the conversion in the sand bed reactor was 0.5 compared with 0.35 without NaOH addition [2]. These test results support the hypothesis that dissolution of silica in the flyash is the limiting step for the reaction between flyash and lime during slurrying. Similar gains in sorbent reactivity were reported when ammonium phosphate or phosphoric acid were used as additives for flyash dissolution [3].

An alternative way of increasing the reaction of $Ca(OH)_2$ with silica is to use a more reactive form of silica. Diatomaceous earth and bentonitic clays were selected for the screening study [4]. Diatomaceous earth or bentonitic clay was reacted with lime by slurrying, and the prepared sorbents were tested in the sand bed reactor. Significant enhancement of reactivity of $Ca(OH)_2$ was possible by using either one of these siliceous materials, with diatomaceous earth being the better silica source. Experimental data indicated that a mixture of diatomaceous earth (MN-53) and CaO at a weight ratio of around 1:1 gives the optimum performance with respect to sorbent conversion with SO_2. This ratio also translates into a molar ratio of SiO_2/CaO of around 1:1.

Another way of increasing the flyash dissolution rate is to increase slurrying temperature. Jozewicz et al. [5] investigated the effects of pressure hydration on the reactivity of the flyash/$Ca(OH)_2$ solids towards SO_2. Pressure-hydrated sorbents with flyash-to-$Ca(OH)_2$ weight ratios ranging from 1:2 to 5:1 were investigated. Results were observed as follows:

- $Ca(OH)_2$ conversion of 0.46 percent at 150 °C, was the maximum observed.
- Conversion of $Ca(OH)_2$ decreased above 150 °C, likely due to formation of cryptocrystalline silicates.
- Conversion of $Ca(OH)_2$ decreased less dramatically as temperatures were lowered from 150 °C to 100 °C.

Another approach to increase the rate of formation of calcium silicates by reacting flyash with $Ca(OH)_2$ is to increase the availability of silica by means of flyash grinding. A laboratory scale grinder using stainless steel balls as a grinding medium was used for grinding. The ash was ground for 1 hr at room temperature and atmospheric pressure. As a result, median particle sized decreased from 12 to 5 μm.

The importance of the weight ratio of siliceous material to $Ca(OH)_2$, hydration time, and hydration temperature, known to have the most significant effects on the reactivity with SO_2 of sorbents prepared with unground flyash and $Ca(OH)_2$, was verified for sorbents produced with ground flyash and $Ca(OH)_2$. The time/temperature effect was tested for hydration time ranging from 1 to 6 hr. Initially, 90 °C hydration temperature was evaluated, and the results are shown in Figure 3. The conversion of $Ca(OH)_2$ in the sand bed reactor increased with increasing hydration time. Conversion increased significantly for the hydration time extended from 1 to 3 hr. However, no significant increase of conversion took place for further increased hydration time from 3 to 6 hr. Reactivities of sorbents produced with the unground flyash are shown for comparison in Figure 3. For the time/temperature conditions tested, higher conversions of $Ca(OH)_2$ were measured in the sand bed reactor for sorbents produced with ground than with unground flyash.

The attractiveness of the furnace sorbent injection process for the removal of SO_2 could be further increased if higher levels of overall sorbent utilization were attainable [6]. One option is to use the unreacted sorbent leaving the boiler to further remove SO_2 downstream. If this sorbent, which under normal conditions is relatively unreactive, could be reactivated to remove SO_2 during its residence time in the duct and particulate control device, it would improve SO_2 capture of the furnace sorbent injection process.

Two sorbents are considered as candidates for injection into the furnace: $CaCO_3$ and $Ca(OH)_2$. Injection of either could result in a mixture of flyash/$CaSO_4$/CaO leaving the boiler. The ratio of $CaSO_4$ to CaO may vary for $Ca(OH)_2$ and $CaCO_3$ injection, depending on their reactivity. To test the potential for reactivation of post-furnace injection solids (PLIS) resulting from the injection of $CaCO_3$ [$CaCO_3$-based PLIS] or $Ca(OH)_2$ [$Ca(OH)_2$-based PLIS], samples were tested for their reactivity with SO_2 following slurrying of "as received" as well as ground solids. The results are presented in Figure 4 giving the reactivity in the form of calcium normalized SO_2 capture in mmol SO_2/g of calcium. Each set of reactivation conditions was tested on $CaCO_3$- and $Ca(OH)_2$-based PLIS. For the reactivation conditions used, there was no significant difference in reactivity of reactivated solids. Grinding with subsequent slurrying at high temperature almost tripled the SO_2 capture, compared to "as received" solids. Grinding appears to be a necessary step in order to reactivate PLIS. Significantly less reactivation of PLIS could be achieved by slurrying of "as received" material.

PROCESS DEVELOPMENT

Bench-scale testing had indicated that lime slurried with silica produced a more reactive sorbent than lime alone. In searching for a way to best utilize this superior sorbent, a series of tests was begun in the 85 m^3/h (50 cfm) pilot facility shown in Figure 5, consisting of a Niro Atomizer spray dryer, a dry sorbent screw feeder, a 5.08 cm (2 in.) I.D. duct varying from 12 to 35 m (40 to 110 ft) in length, a cyclone module, and a pulse-jet baghouse having a 2.3 m^2 (25 ft^2) collection area. Electrically heated air augmented with SO_2 (typically at 1,500-2,000 ppmv) serves as a surrogate flue gas [4].

Initial tests, in March 1986, examined the sorbent slurry prepared by mixing 5 parts flyash, 1 part $Ca(OH)_2$, and 14 parts water in a heated tank at 90 °C for 12-16 hr. The slurry then was injected via a spray dryer into a gas stream. Although 40-50 percent SO_2 removal was observed, severe wall deposits were noted at approaches to saturation of 10-50 °C. Subsequently, tests were conducted using spray dried sorbent prepared similarly to that above, but injected as a powder into a prehumidified gas stream of 11-17 °C approach to saturation. With this approach a 50 percent SO_2 removal at Ca/S=1.0 and 80 percent at Ca/S=2.0 were observed across the baghouse. After this time (April 1986), all subsequent process development focused on a dry sorbent [4,7].

In September 1986, spray dried silicates prepared at 90 °C for 8 hr were shipped to the Electric Power Research Institute's (EPRI's) Arapaho pilot facilities for evaluation. Despite the fact that the "EPA hydrate" fared better than any EPRI calcium-based material—94 percent and 55 percent SO_2 removal at Ca/S = 2.0 and ΔT_{sat} = 11 °C for a fabric filter and electrostatic precipitator collector, respectively—the performance was not as good as EPRI's results for sodium sorbents [8]. Examination of the sorbent shipped to Arapaho showed a significant deterioration (via an unknown mechanism) in surface area (from 46 to 31 m^2/g) due to spray drying prior to shipment or change during transport. Therefore, in October 1986, other methods of converting silicate slurry to a dry sorbent were investigated.

An alternative method to thermal drying was developed wherein a slurry of flyash and hydrated lime was reacted at a ratio of 3:1 for 12 hr, and filtered to obtain a cake of 30-60 percent moisture. A cement mixer was used to blend this cake with dry recycle solids produced from a previous test run. The 3 parts of dry recycle solids combined with 1 part of wet cake produced a damp powder containing 25-33 percent moisture [7]. At this point, a second important property of calcium silicates became

evident—that of moisture-carrying ability. The 25-33 percent moisture solids generated by mixing slurry or filter cake with dry recycle solids were generally free-flowing and dry to the touch. Any lumps in the mixture could be easily pulverized by modest shaking or even drying (when injected). With the development of more reactive calcium silicates, this moisture-carrying ability has been shown to increase in proportion to surface area; therefore, the reactivity toward SO_2 increases with increased moisture content.

In 1987, EPA and Acurex began preparing pressure hydrated sorbents in 50 percent solids slurries and mixing 2 parts dry recycle solids with 1 part fresh slurry [9]. The pilot plant results for silicates injected into a humidified gas stream showed typically 50 percent SO_2 removal in-duct (for 2 s residence time) and 90 percent SO_2 removal through the fabric filter at a Ca/S ratio of 1.0 as shown in Figure 6.

In 1988, EPA and Acurex made an extensive set of runs using furnace injected sorbent waste as the starting material for calcium silicate. The results showed that up to 80 percent additional capture of SO_2 could be achieved at a recycle ratio of 2.0 using furnace solids from a furnace injection (LIMB) process that was 25 percent utilized in the furnace (50 percent SO_2 capture at Ca/S = 2.0)[10]. In this evaluation, the newly evolved process was christened ADVACATE for advanced silicate.

In April-May 1989, EPA sponsored a field pilot evaluation of a combined LIMB-ADVACATE process where a 105 MW_e LIMB demonstration (Edgewater Plant, Lorain, OH) fed a 3400 m^3/hr (2,000 cfm) slipstream through a pilot ESP. The results of this evaluation, shown in Figure 7 and Table 1, were so encouraging–up to 98 percent SO_2 removal–that intense efforts regarding possible demonstrations were begun [11]. It is worth noting here that current sorbents are roughly three times more reactive than those prepared and used at Edgewater.

Referring to Figure 6, AEERL was negotiating the first EPA Cooperative Research Agreement under the Federal Technology Transfer Act of 1986 with Flakt, USA, of Knoxville, TN, during 1987-88 when it was learned that Flakt was developing its own dry injection technology, called Moist Dust Injection or MDI. Flakt's concept was to create a dense-phase reaction in-duct using moistened recycle product with fresh lime to simultaneously capture SO_2 and humidify the gas in the duct.

Following the signing of this agreement in November 1988, silicate sorbent was prepared at a 3:1 flyash-to-lime ratio at about 20 percent moisture for Flakt to use in place of lime in their MDI pilot plant at the University of Tennessee in Knoxville. In early December 1988, Flakt conducted runs at 1000, 2000, and 3000 ppmv SO_2 at a ΔT_{sat}=11 °C and realized SO_2 removals of ~80 percent in-duct and 96 percent through the fabric filter at a Ca/S=1.0 as shown in Figure 6. The reason for this improvement in in-duct SO_2 capture—80 percent at Flakt versus 50 percent at the EPA pilot plant located at Research Triangle Park (RTP), NC—is still uncertain. However, in recent months, attempts to simulate MDI at RTP have shown up to 60 percent SO_2 capture at Ca/S=1.0. Currently the effect of gas-phase turbulence (Reynolds number) on SO_2 capture in-duct is being studied as a possible explanation. For the time being, it is sufficient to know that SO_2 capture using MDI/ADVACATE appears to improve with increasing duct dimensions, assuming an adequate duct velocity for good mixing (at least 15 m/s).

As a final note on process development, silicate sorbents of 90 m^2/g are routinely produced at RTP in 2-3 hr and show an ability to hold 60 percent moisture and be free-flowing. Such a remarkable improvement in sorbent characteristic suggests the need to update previous pilot plant data. A more intriguing notion is operation of dust collectors at saturation temperatures. If a silicate sorbent holds up to 60 percent moisture and appears dry, is condensation even feasible for a cooled gas-stream containing dry or partially dried silicates? By the time this paper is presented in August 1990, the authors should have more insight into operation at apparent saturation conditions.

The impact of an ADVACATE, LIMB-ADVACATE, or ADVACATE/MDI process upon particle collection has been ignored up to now because we have little experience with ADVACATE-ESP operation, although it is acknowledged that ESPs do not normally operate at high dust loadings. Conversely, both EPA and Flakt have considerable experience with ADVACATE and MDI processes on fabric filters, and no significant operational problems have occurred.

CURRENT STATUS

Figure 8 represents the diverse process schemes for ADVACATE. The source of calcium for silicates may be furnace-injected lime or limestone, lime added to the silicate slurry tanks, or, in the case of many Western coals, calcium occurring in coal ash. Waste solids from a LIMB or fluid-bed combustor (FBC) are also a potential economic source of calcium.

The way sorbent is injected depends upon vendor preference and calcium source. In LIMB experience, limestone or lime is pneumatically conveyed into the furnace while ADVACATE sorbent has been fed via direct screw feeding, pneumatic conveying, or the proprietary MDI process of Flakt. The flue gas may be cooled/humidified via moisture injection, indirect gas cooling, evaporation of moisture in damp solids injected, or a combination of these techniques. Particle collection is either by fabric filter or ESP, although the latter may require a precollector to protect electrical integrity, as well as segregate the coarser (less reacted) material for recycle.

EPA's current plans include a 10 MW_e field pilot evaluation at the Shawnee No. 9 facility of the Tennessee Valley Authority (TVA) starting in 1991. Several sources of sorbent, including FBC and LIMB waste, will be evaluated on a 2.5-3.0 percent sulfur Midwestern coal.

If successful, the Shawnee effort will be followed by augmentation of the LIMB demonstration at Virginia Power's Yorktown No. 2 (180 MW_e) unit starting in 1992. This effort currently is scheduled for installation of LIMB using a hydrated

lime sorbent, but can be converted to a furnace-injected limestone-ADVACATE process upstream of an ESP. A 2.4 percent sulfur Eastern bituminous coal will be fired at Yorktown for this demonstration.

Another interesting process variation is the ability of ADVACATE to use the natural alkalinity of Western coal flyash to remove SO_2. In many cases, such as Powder River subbituminous coal, a variation of ADVACATE can potentially remove all the SO_2 at roughly half the cost as presented later in this paper. This is because half the cost of ADVACATE is for lime or limestone receiving, storage, and handling. The low-sulfur ADVACATE process variation would also incur no additional solid waste (except for the weight of sulfite/sulfate added due to SO_2 absorption). This option is slated for study later in 1990 on a pilot scale at RTP.

COST ESTIMATES

Based on the preliminary design of an ADVACATE/MDI process for the Yorktown demonstration, cost estimates have been prepared by Radian Corporation using EPRI economic premises. Performance predictions based on the Flakt field evaluation for ADVACATE and Edgewater results for LIMB/ADVACATE have been incorporated along with cost estimates. In Figure 9 the total annualized cost estimates for a 300 MW_e retrofit using 2.2 percent sulfur coal and the furnace limestone ADVACATE/MDI system are compared with wet FGD. Although the economic assumptions are too numerous to discuss here, the basic philosophy is that all factors are comparable except that the highest process and project contingency factors have been assigned to ADVACATE due to the unproven nature of the technology.

For utility-scale applications, the estimated cost of ADVACATE is typically 30-35 percent capital and 40-50 percent operating of the costs for wet FGD. Table 2 shows a similar analysis performed on slurry costs of wet FGD vs. lime and limestone ADVACATE for a 350 MW_e, 2.2 percent sulfur coal retrofit. ADVACATE is projected to cost around $60/kW and, for this case, $225-250/ton SO_2 removed, compared with $195/kW and $520/ton SO_2 removed for wet FGD. In these comparisons, the wet FGD was assumed to have no spare modules, no reheat, and a retrofit factor of 1.5.

PROCESS SUMMARY

The key feature of the ADVACATE process is its unique calcium silicate sorbent which has the following properties:

- Typical surface area of 70-90 m^2/g.
- Reactivity toward SO_2 4-6 times that of lime.
- Ability to carry high (60 percent) moisture content and be free flowing.

High calcium utilization observed in bench, pilot, and field pilot evaluations is likely due to the free calcium's being all surface calcium. Unlike conventional sorbent injection technology, spent ADVACATE sorbent does not have an unreacted core.

The process developed for this sorbent has the following features:

- No absorber vessel.
- No requirement for separate humidification of flue gas.
- Dry waste solids.
- Various plant waste waters, including boiler blowdown, can be used for ADVACATE slurry.
- SO_2 removal comparable to wet FGD at less than half the cost.

Additionally, virtually no potential for in-duct sorbent deposits exists due to the hygroscopic nature of ADVACATE sorbent. Potential operation at apparent saturation temperatures seems feasible.

At this time, the process has not operated in a continuous recycle mode, nor has ESP performance with ADVACATE injection been studied in depth. These and other questions should be resolved at Shawnee during 1991.

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Table 1. Edgewater SO_2 Removal at Ca/S = 2.0, H_{out} = 63 °C

Dry ADVACATE Injection Rate (g/dscm)	Total SO_2 Removal (%)	SO_2 Removal ADVACATE Only (%)
0	65	0
10	84	54
20	93	80
30	97	91
40	99	97

Table 2. ADVACATE Retrofit Economic Evaluation
(Basis: 350 MWe, 2.23% S coal, May 1988 dollars, limestone=$9.76/tonne, lime=$67.46/tonne)

	Wet FGD	Limestone/ ADVACATE	Lime/ ADVACATE
Capital cost, $/kW	195	58	59
Cost, $/tonne SO_2	520	224	255

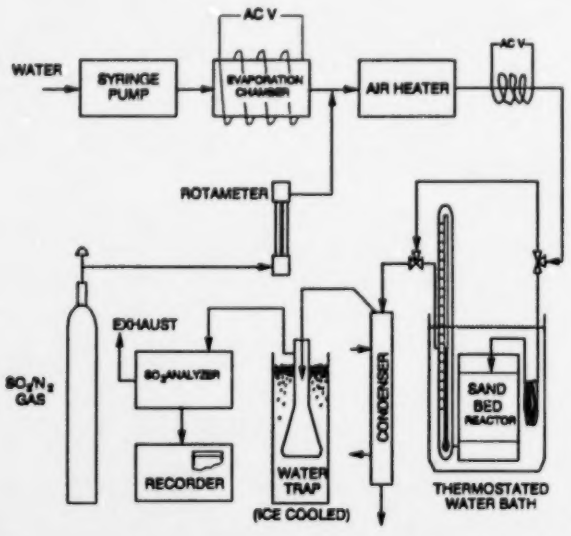

Figure 1. Bench-scale sand bed reactor.

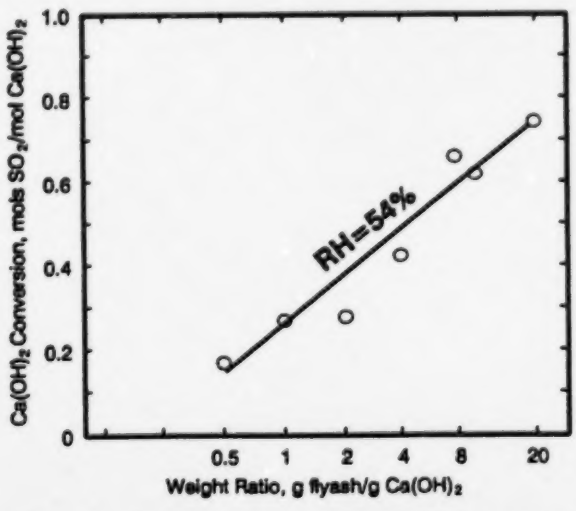

Figure 2. The effect of flyash-to-$Ca(OH)_2$ weight ratio on $Ca(OH)_2$ conversion (54 percent relative humidity, 500 ppm SO_2).

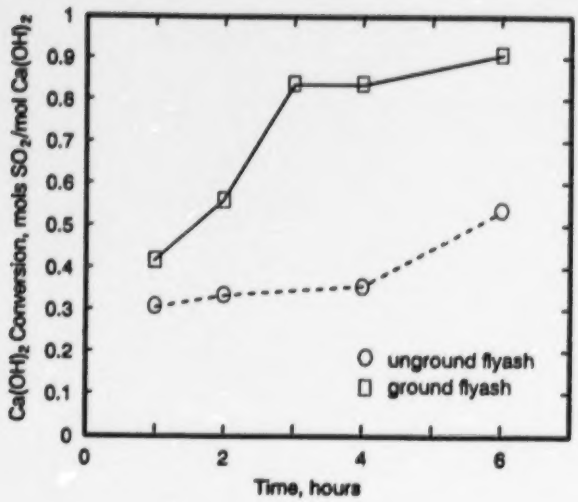

Figure 3. The effect of slurrying time on the conversion of $Ca(OH)_2$ (90 °C slurring, flyash-to-$Ca(OH)_2$ weight ratio of 3:1).

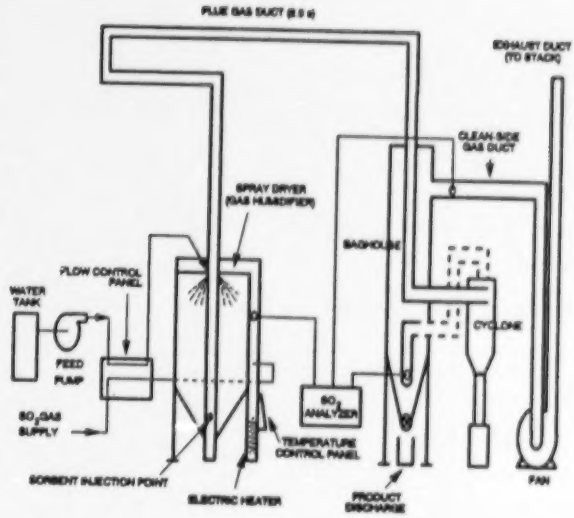

Figure 5. Pilot plant facility.

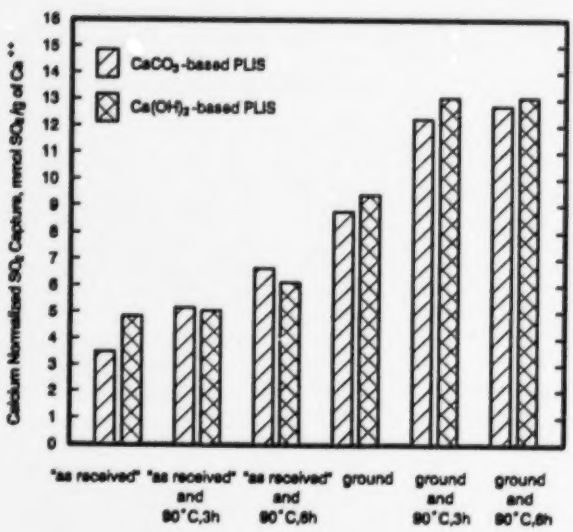

Figure 4. Comparative reactivity evaluation of $CaCO_3$- and $Ca(OH)_2$-based PLIS.

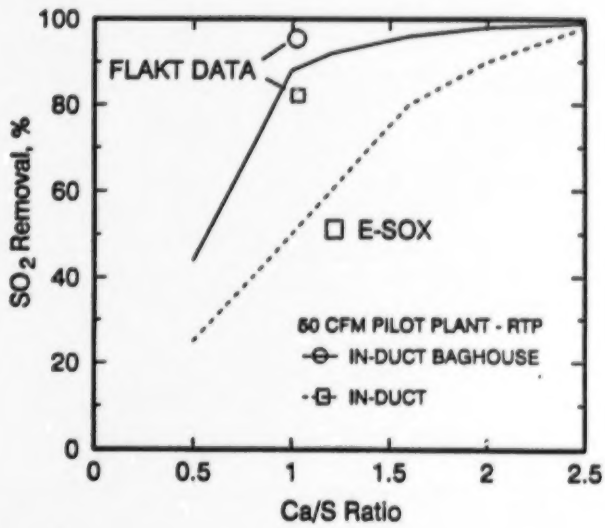

Figure 6. SO_2 removal vs. stoichiometric ratio for calcium silicate damp injection.

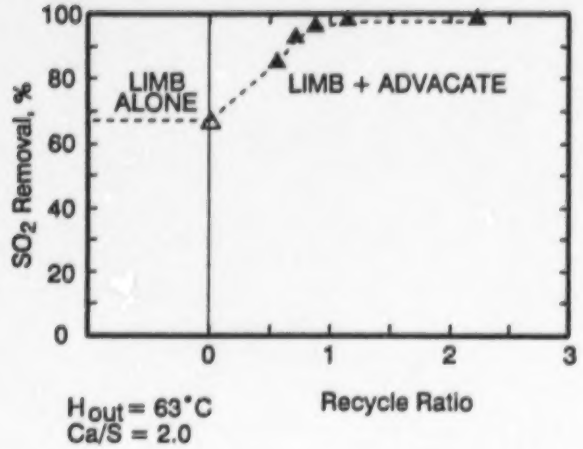

Figure 7. Edgewater LIMB/ADVACATE results.

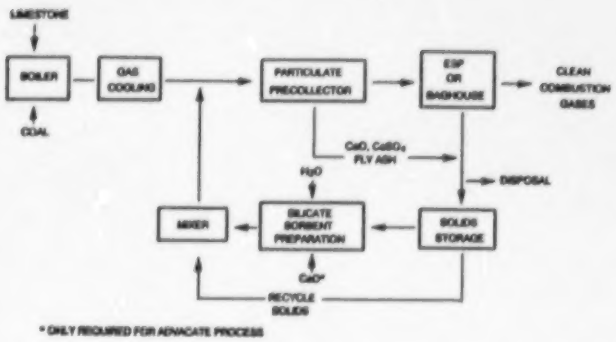

Figure 8. ADVACATE process schematic.

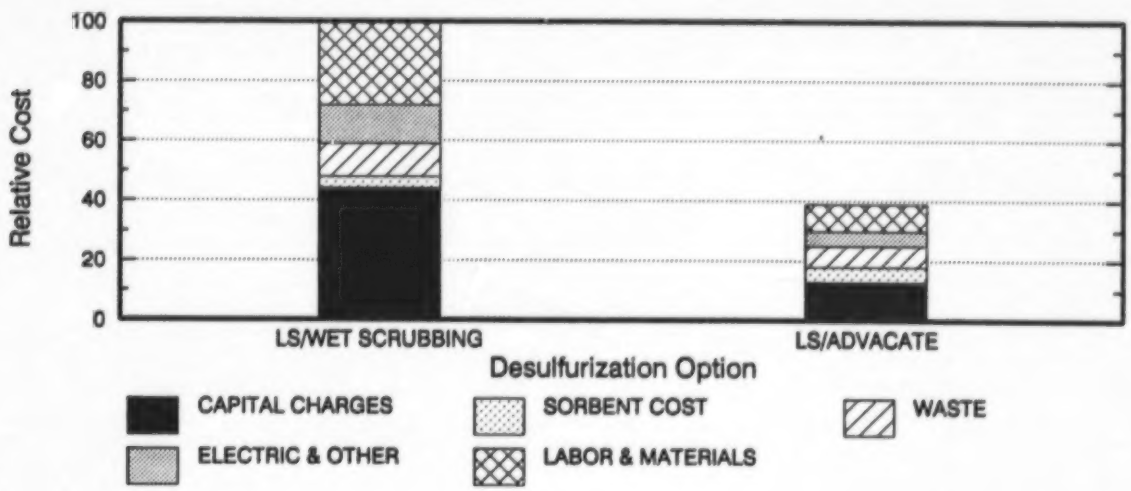

Figure 9. Total cost comparison - limestone wet scrubbing vs. limestone/ADVACATE.

This paper has been reviewed in accordance with the U.S. Environmental Protection Agency's peer and administrative review policies and approved for presentation and publication.